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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

Ting Tao, et al

PROCESS FOR THE PREPARATION
OF CYANINE DYE WITH
POLYSULFONATE ANIONS

Serial No. 10/722,257

Filed 25 November 2003

Publication No. 2005/0113546 A1
Published May 26, 2005

Commissioner for Patents
P.O. Box 1450
Alexandria, VA. 22313-1450

Sir:

**REQUEST FOR CORRECTION TO PATENT
APPLICATION PUBLICATION**

A material mistake has been discovered in the above-captioned Publication. An incorrect set of claims has been published (copy attached). Enclosed with this Request are the correct claims 1-19.

Therefore, we are requesting a correction to the patent application Publication due to a United States Patent and Trademark Office error.

The Commissioner is hereby authorized to charge any fees in connection with this communication to Eastman Kodak Company Deposit Account No. 05-0225. **A duplicate copy of this letter is enclosed.**

Respectfully submitted,

Attorney for Applicant(s)
Registration No. 27,678

J. Lanny Tucker/s-p
Rochester, NY 14650
Telephone: (585) 722-9332
Facsimile: (585) 477-1148

If the Examiner is unable to reach the Applicant(s) Attorney at the telephone number provided, the Examiner is requested to communicate with Eastman Kodak Company Patent Operations at (585) 477-4656.

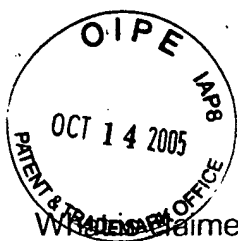
Group Art Unit: 1752

Examiner:

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Sherry A. Payne

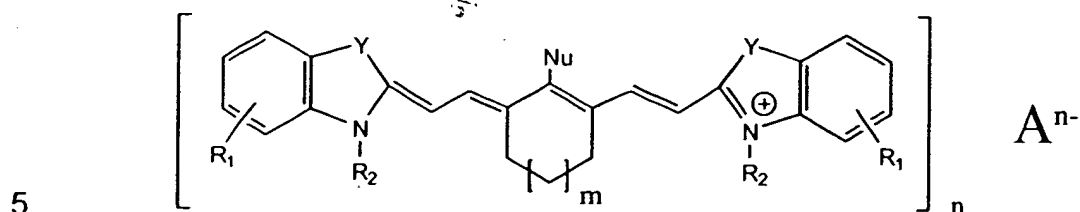
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CLAIMS

What is claimed is:

1. A method for preparing infrared absorbing cyanine dyes of the structure:



in which:

R_1 is hydrogen, or R_1 is one or more alkyl, alkoxy, carboxyl, nitro, cyano, trifluoromethyl, acyl, alkyl or aryl sulfonyl, or halogen groups, or R_1 is the atoms necessary to form a substituted or unsubstituted benzo group;

10 R_2 is alkyl, aryl, or aralkyl;

Nu is halogen, substituted or unsubstituted phenoxy, substituted or unsubstituted thiophenoxy, or substituted or unsubstituted diphenylamino;

Y is O, S, NR' , or $C(R')_2$, where R' is hydrogen or alkyl;

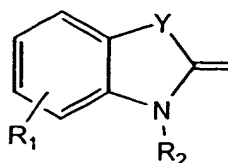
m is zero or one;

15 n is two, three, or four; and

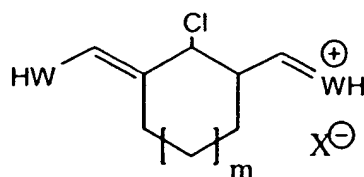
A is an aromatic group that has n sulfonate groups;

the method comprising the steps of:

a) reacting an activated methylene group containing a heterocyclic base of the structure:



20 with a compound of the structure:



and forming an intermediate in a reaction mixture;

in which W is O or Ar-N, Ar is an aromatic group, X⁻ is an anion, and m is zero or one, and

b) adding a salt of A to the reaction mixture; and

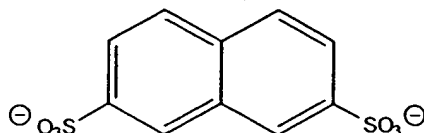
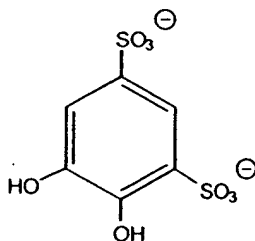
5 c) isolating the infrared absorbing cyanine dye;

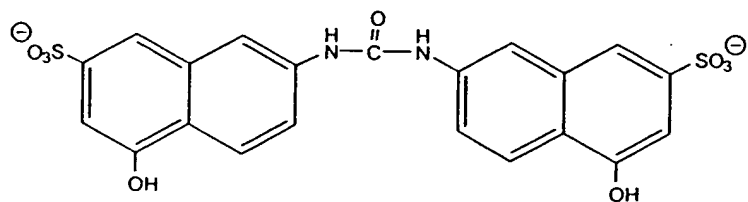
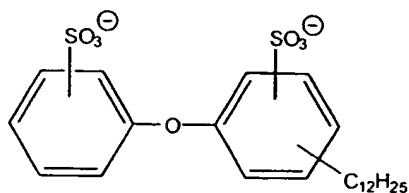
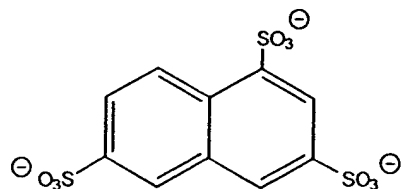
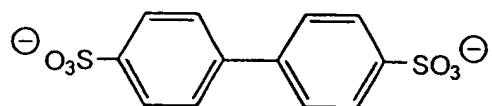
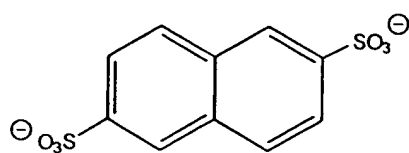
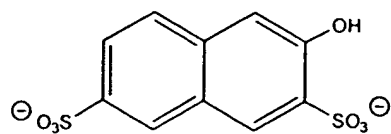
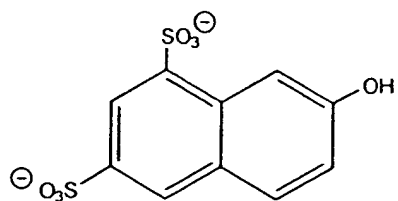
in which the infrared absorbing cyanine dye is the only compound isolated in the method.

2. The method of claim 1 in which R₁ is hydrogen; R₂ is methyl, ethyl, *n*-propyl, or *n*-butyl; Nu is chloro, phenoxy, thiophenoxy, or diphenyl amino; and

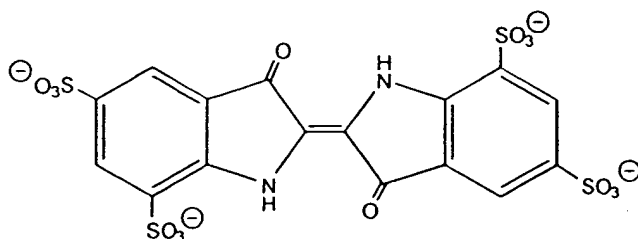
10 Y is C(CH₃)₂, O, or S.

3. The method of claim 2 in which A is selected from the group consisting of biphenyl-4,4'-disulfonate; diphenyl ether-4,4'-disulfonate; stilbene-2,2'-disulfonate; 2,2'-dihydroxy-4,4'-dimethoxybenzophenone-5,5'-disulfonate,





, and



4. The method of claim 2 in which n is 2 and A is 4,5-dihydroxy-1,3-benzenedisulfonate.

5. The method of claim 1 in which Nu is chloro.

5 6. The method of claim 5 in which R_1 is hydrogen; R_2 is methyl, ethyl, n -propyl, or n -butyl; and Y is $C(CH_3)_2$, O , or S .

7. The method of claim 6 in which n is 2 and A is 4,5-dihydroxy-1,3-benzenedisulfonate.

8. The method of claim 7 in which Y is $C(CH_3)_2$.

10 9. The method of claim 8 in which n is 2 and A is 4,5-dihydroxy-1,3-benzenedisulfonate.

10. The method of claim 1 additionally comprising, after step a) and before step b), an additional step of adding a substituted or unsubstituted phenoxy, a substituted or unsubstituted thiophenoxy, or a substituted or unsubstituted diphenylamino compound to the reaction mixture.

11. The method of claim 10 in which R_1 is hydrogen; R_2 is methyl, ethyl, n -propyl, or n -butyl; Nu is phenoxy, thiophenoxy, or diphenyl amino; and Y is $C(CH_3)_2$, O , or S .

12. The method of claim 11 in which A is 4,5-dihydroxy-1,3-benzenedisulfonate.

13. The method of claim 12 in which Y is $C(CH_3)_2$.

14. The method of claim 13 in which n is 2 and A is 4,5-dihydroxy-1,3-benzenedisulfonate.

15. The method of claim 1 in which the dye is isolated by filtration.

16. The method of claim 15 in which R_1 is hydrogen; R_2 is methyl, ethyl, *n*-propyl, or *n*-butyl; Nu is chloro, phenoxy, thiophenoxy, or diphenyl amino; and Y is $C(CH_3)_2$, O, or S.

5 17. The method of claim 16 in which A is 4,5-dihydroxy-1,3-benzenedisulfonate.

18. The method of claim 1 in which:

R_1 is hydrogen; R_2 is methyl, ethyl, *n*-propyl, or *n*-butyl; Nu is chloro; Y is $C(CH_3)_2$, O, or S; and A is 4,5-dihydroxy-1,3-benzenedisulfonate; and

10 the method consists essentially of steps a), b), and c).

19. The method of claim 1 in which:

R_1 is hydrogen; R_2 is methyl, ethyl, *n*-propyl, or *n*-butyl; Nu is phenoxy, thiophenoxy, or diphenyl amino; R_2 is methyl, ethyl, *n*-propyl, or *n*-butyl; Y is $C(CH_3)_2$, O, or S; and A is 4,5-dihydroxy-1,3-benzenedisulfonate; and

15 the method consists essentially of steps a), b), c), and an additional step of adding a substituted or unsubstituted phenoxy, a substituted or unsubstituted thiophenoxy, or a substituted or unsubstituted diphenylamino compound to the reaction mixture.

EXAMPLE 7

[0080] This example illustrates the one pot synthesis of 2-[2-[2-chloro-3-[(1,3-dihydro-1,3,3-trimethyl-2H-indol-2-ylidene)ethylidene]-1-cyclohexen-1-yl]ethenyl]-1,3,3-trimethyl-3H-Indolium, salt with 4,5-dihydroxy-1,3-benzenedisulfonic acid (2:1) (Compound 1) from intermediate A, without a catalyst.

[0081] In a 250 ml flask equipped with a stirring bar and a condenser attached to a Dean-Stark trap, 11.8 g of Fisher's base, 3.45 g of intermediate A, 5.0 g of 36.5% hydrochloric acid, 50 ml of benzene and 100 ml of n-propanol were heated at reflux in an oil bath heated at 100° C. for 3 h. After 3 h, no further water was collected in the Dean-Stark trap. The reaction flask was attached to a rotary evaporator and the solvents evaporated. The residual green material was dissolved in 150 g of ethanol and 150 g of water was added. The resulting dark-green solution was then added drop-wise to a solution containing 10 g of disodium 4,5-dihydroxy-1,3-benzenedisulfonic acid (from Aldrich) in 50 g of water and 50 g of ethanol while stirring. The resulting precipitate was collected by filtration, washed with 2x50 ml of ethyl acetate, and dried at ambient temperature overnight. Yield: 11.5 g.

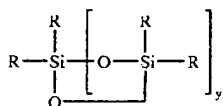
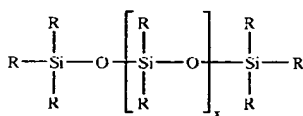
[0082] Proton NMR (in DMSO-d₆): delta: 1.66 (24H, s), 1.86 (4H, br), 2.71 (8H, t), 3.68 (12H, s), 6.30 (4H, d), 6.97 (1H, s), 7.20-7.35 (5H, m), 7.35-7.50 (8H, m), 7.62 (4H, d), 8.25 (4H, d), 8.88 (1H, s), and 10.60 (1H, s).

[0083] Having described the invention, we now claim the following and their equivalents.

Incorrect Claims

1-5. (canceled)

6. A process for the production of siloxane oligomers of the general formulae I or II



in which x is an integer from 0 to 1000, y is a number from 1 to 1000, and the substituents R are identical or different and consist of functionalised alkyl groups, (C₁-C₁₈)alkyl, (C₁-C₄)alkoxy, (C₁-C₄)haloalkoxy, phenyl, aryl, aralkyl or hydroxy groups, wherein at least one functionalised alkyl group is present per oligomer molecule, the method comprising:

treating a halogenalkyltrihalogenasilane to oligomerization in the presence of alcohol and water and co-oligomerized with at least one of a (C₁-C₁₈)-alkyl-, phenyl-, aryl- or aralkyl-trihalogenasilane and silicon tetrachloride, optionally modifying a halogenalkyl function in a further step.

7. The process for the production of the siloxane oligomer according to claim 6, further comprising modifying the halogenalkyl function with ammonia and separating ammonium halide.

8. The process for the production of the siloxane oligomer according to claim 6, further comprising modifying the halogenalkyl function with sodium methacrylate or potassium methacrylate and separating sodium halide or potassium halide.

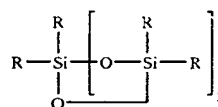
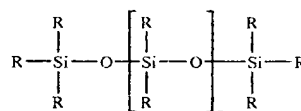
9. The process for the production of the siloxane oligomer according to claim 6, further comprising modifying the halogenalkyl function with ammonia and hydrogen sulfide or ammonium hydrogen sulfide and separating ammonium halide, or modifying with sodium hydrogen sulfide or potassium hydrogen sulfide separating sodium halide or potassium halide.

10. The process for the production of the siloxane oligomer according to claim 6, further comprising modifying the halogenalkyl function with sodium, potassium or ammonium rhodanide and separating sodium, potassium or ammonium halide.

11. The process for the production of the siloxane oligomer according to claim 6, further comprising modifying the halogenalkyl function with sodium azide and separating sodium halide.

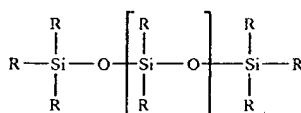
12. The process for the production of the siloxane oligomer according to claim 6, further comprising modifying the halogenalkyl function with sodium polysulfide or with sodium sulfide and sulfur or sodium polysulfide and sodium sulfide, and separating sodium halide.

13. A rubber composition containing a siloxane oligomer of the general formulae I or II

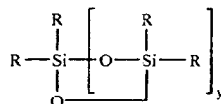


in which x is an integer from 0 to 1000, y is a number from 1 to 1000, and the substituents R are identical or different and consist of functionalised alkyl groups, (C₁-C₁₈)alkyl, (C₁-C₄)alkoxy, (C₁-C₄)haloalkoxy, phenyl, aryl, aralkyl or hydroxy groups, wherein at least one functionalised alkyl group is present per oligomer molecule as a coupling agents.

14. A rubber composition, comprising rubber, at least one of a precipitated silica and carbon black, and a siloxane oligomer of the general formulae I or II



-continued

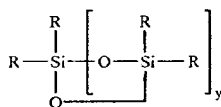
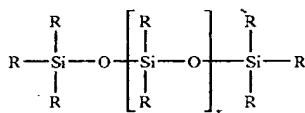


in which x is an integer from 0 to 1000, y is a number from 1 to 1000, and the substituents R are identical or different and consist of functionalised alkyl groups, (C₁-C₁₈)alkyl, (C₁-C₄)alkoxy, (C₁-C₄)haloalkoxy, phenyl, aryl, aralkyl or hydroxy groups, wherein at least one functionalised alkyl group is present per oligomer molecule.

15. The rubber composition according to claim 14 wherein the rubber is polybutadien, polyisoprene, styrene/butadiene copolymers with styrene content of 1 to 60 wt. %, isobutylene/isoprene copolymers, butadiene/acrylonitrile copolymer with acrylonitrile content of 5 to 60 wt. %, ethylene/propylene/diene copolymer or mixtures of these rubbers.

16. The rubber composition according to claim 14 further comprising at least one of a reaction accelerator, reaction retarder, anti-ageing agent, stabilizer, processing auxiliary, plasticizer, wax, metal oxide, and activator.

17. A process for making a rubber composition comprising mixing a rubber with a siloxane oligomer of the general formulae I or II



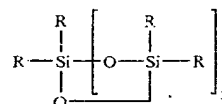
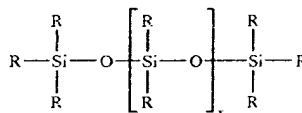
in which x is an integer from 0 to 1000, y is a number from 1 to 1000, and the substituents R are identical or different and consist of functionalised alkyl groups, (C₁-C₁₈)alkyl, (C₁-C₄)alkoxy, (C₁-C₄)haloalkoxy, phenyl, aryl, aralkyl or hydroxy groups, wherein at least one functionalised alkyl group is present per oligomer molecule,

a filler and optionally a rubber auxiliary substance in at least one thermomechanical mixing stage at 100 to 170° C., and

adding the resulting mixture to an internal kneader or roller at 40 to 110° C. together with a crosslinking agent.

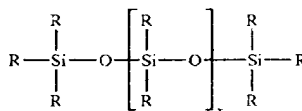
18. The process according to claim 17 further comprising shaping the resulting rubber composition into the desired article and vulcanizing to obtain a vulcanized rubber article.

19. A rubber tire containing a siloxane oligomer of the general formulae I or II



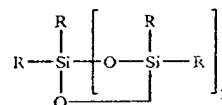
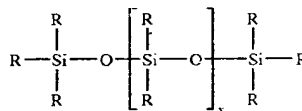
in which x is an integer from 0 to 1000, y is a number from 1 to 1000, and the substituents R are identical or different and consist of functionalised alkyl groups, (C₁-C₁₈)alkyl, (C₁-C₄)alkoxy, (C₁-C₄)haloalkoxy, phenyl, aryl, aralkyl or hydroxy groups, wherein at least one functionalised alkyl group is present per oligomer molecule.

20. A shaped rubber article containing a siloxane oligomer of the general formulae I or II



in which x is an integer from 0 to 1000, y is a number from 1 to 1000, and the substituents R are identical or different and consist of functionalised alkyl groups, (C₁-C₁₈)alkyl, (C₁-C₄)alkoxy, (C₁-C₄)haloalkoxy, phenyl, aryl, aralkyl or hydroxy groups, wherein at least one functionalised alkyl group is present per oligomer molecule.

21. A process for the production of siloxane oligomers of the general formulae I or II



in which x is an integer from 0 to 1000, y is a number from 1 to 1000, and the substituents R are identical or different and consist of functionalised alkyl groups, (C₁-C₁₈)alkyl, (C₁-C₄)alkoxy, (C₁-C₄)haloalkoxy, phenyl, aryl, aralkyl or hydroxy groups, wherein at least one functionalised alkyl group is present per oligomer molecule.

hydroxy groups, wherein at least one functionalised alkyl group is present per oligomer molecule, the method comprising:

treating a halogenalkyltrihalogensilane to oligomerization in the presence of alcohol and water and optionally co-oligomerized with at least one of a (C₁-C₁₈)-alkyl-, phenyl-, aryl- or aralkyl-trihalogensilane and silicon tetrachloride, and

modifying the halogenalkyl function with:

- a) ammonia and separating ammonium halide,
- b) sodium methacrylate or potassium methacrylate and separating sodium halide or potassium halide,

c) ammonia and hydrogen sulfide or ammonium hydrogen sulfide and separating ammonium halide,

d) sodium hydrogen sulfide or potassium hydrogen sulfide and separating sodium halide or potassium halide,

e) sodium, potassium or ammonium rhodanide and separating sodium, potassium or ammonium halide,

f) sodium azide and separating sodium halide, or

g) sodium polysulfide, sodium sulfide and sulfur, or sodium polysulfide and sodium sulfide, and separating sodium halide.

* * * * *